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权利要求书 1 页 说明书 18 页 附图页数 6 页

[54]发明名称 一种降低汽油烯烃、硫和氮含量的催化转化方法

[57]摘要

一种降低汽油烯烃、硫和氮含量的催化转化方法,是将预热后的汽油原料进入提升管或流化床反应器内与催化剂接触,在反应温度为 100~600℃、重时空速为 1~120 小时⁻¹、催化剂与汽油原料的重量比为 2~15:1、水蒸汽与原料油的重量比为 0~0.10:1 的条件下进行催化转化反应,分离反应产物,待生催化剂经汽提、再生、冷却后返回到反应器循环使用。采用本发明提供的方法,汽油组成中的烯烃可以降低到20 重%,汽油组成中的异构烷烃可以增加到66 重%。

权 利 要 求 书

1、一种降低汽油烯烃、硫和氮含量的催化转化方法，其特征在于将预热后的汽油原料进入提升管或流化床反应器内与催化剂接触，在反应温度为 100～600℃、进料重时空速为 1~120 小时⁻¹、催化剂与汽油原料的重量比为 2~15:1、水蒸汽与原料油的重量比为 0～0.1:1 的条件下进行催化转化反应，分离反应产物，待生催化剂经汽提进入再生器再生，再生催化剂冷却后返回反应器循环使用。

2、按照权利要求 1 所述的催化转化方法，其特征在于该方法既可以在单独的提升管或流化床反应器中进行，也可以在与另一个提升管或流化床反应器联合的装置上实施。

3、按照权利要求 1 所述的催化转化方法，其特征在于所述的再生催化剂冷却是在冷却器中进行的，该冷却器可以与再生器连为一体，也可以通过管线与再生器相连。

4、按照权利要求 1 所述的催化转化方法，其特征是所述催化剂的活性组分选自含或不含稀土的 Y 或 HY 型沸石、含或不含稀土的超稳 Y 型沸石、ZSM-5 系列沸石或具有五元环结构的高硅沸石、β 沸石、镁碱沸石中的一种、两种或三种。

5、按照权利要求 1 所述的催化转化方法，其特征是所述催化剂可以是无定型硅铝催化剂。

6、按照权利要求 1 所述的催化转化方法，其特征是所述的汽油原料中的烯烃含量为 20～90 重%。

7、按照权利要求 1 所述的催化转化方法，其特征在于反应温度为 150～550℃，进料重时空速为 2~100 小时⁻¹，催化剂与汽油原料的重量比为 3~10:1，水蒸汽与汽油原料的重量比为 0.01~0.05:1。

说 明 书

一种降低汽油烯烃、硫和氮含量的催化转化方法

本发明属于石油烃类的催化转化方法，具体地说，是属于降低汽油组成中的烯烃含量、硫和氮含量的催化转化方法。

随着环境对汽油产品的质量要求日益严格，汽油已由无铅汽油发展到新配方汽油，新配方汽油的标准是要求限制汽油的蒸气压和苯含量，还将逐步限制芳烃和烯烃含量，世界各国和地区制定的新配方汽油标准不尽相同，但一个总的原则是汽油组分中的烯烃含量在 20 重％以下，硫含量在 100ppm以下。

目前商品汽油组分中烯烃的含量在 35~65 重％，富含烯烃的汽油热稳定性差，在汽车行驶过程中烯烃在内燃机燃油系统的温度条件下会氧化成沉积物附着在化油器的主油量口、怠速油量孔的内壁和节气门的表面，很快导致在用汽车怠速不稳、提速供油不畅、汽油燃烧不充分，使碳氢化合物、一氧化碳排放量增加，污染环境。此外，烯烃蒸发到大气中同臭氧结合形成毒性化合物。

另一方面，由于原料油的不断变重，商品汽油中的硫含量也在增加，商品汽油中的硫含量大约在 300ppm 以上。对于这类汽油必须先经加氢精制，才能满足新配方汽油的质量要求，但该方法加工流程复杂并且经济上不合理。

本发明的目的是提供一种能将汽油中的烯烃催化转化为异构烷烃和芳烃并且能降低其硫、氮含量的方法。

本发明的目的是通过下述方案达到的：预热后的汽油原料进入提升管或流化床反应器内，在或不在水蒸汽存在下与催化剂接触，反应产物、水蒸汽和反应后带焦炭的待生催化剂进行气固分离，分离反应产物得到汽油产品和少量的干气、液化气、柴油，待生催化剂经水蒸汽汽提后输送到再生器，在含氧气体存在下进行烧焦再生，热的再生催化剂经冷却后返回反应器循环使用。

本发明的具体操作步骤如下：预热后的汽油原料进入提升管或流化床反应器内与催化剂接触，在反应温度为 100~600℃，最好为 150~550℃、反应压力为120~150千帕，最好为250~400千帕，总重量空速为1~120小时[-1]、

1

比（以下简称水油比）为 0～0.1:1，最好为 0.01～0.05:1 的条件下进行异构化反应和氢转移反应，反应产物、水蒸汽和反应后带焦炭的待生催化剂进行气固分离；分离反应产物得到干气、富含丙烯和异丁烷的液化气、富含异构烷烃和芳烃的汽油及柴油；待生催化剂进入汽提器，用水蒸汽汽提出催化剂上吸附的烃类产物；然后待生催化剂输送到再生器，在含氧气体存在下进行烧焦再生；热的再生催化剂经冷却器冷却后返回反应器循环使用。

本发明所用的汽油原料组成中的烯烃含量为 20～90 重%，硫含量在 200ppm 以上；该汽油馏分可以是全馏分，也可以是其中某一段窄馏分。

本发明的产物是汽油产品和少量的气体、柴油。其中汽油产品中烯烃、硫和氮含量较低，而异构烷烃和芳烃含量较高；液化气中富含丙烯和异丁烷。

本发明适用的催化剂可以是活性组分选自含或不含稀土的 Y 或 HY 型沸石、含或不含稀土的超稳 Y 型沸石、ZSM-5 系列沸石或具有五元环结构的高硅沸石、β沸石、镁碱沸石中的一种、两种或三种的催化剂，也可以是无定型硅铝催化剂。

本发明所用的提升管或流化床反应器可以是常规的提升管或流化床反应器，也可以是在常规的提升管或流化床基础上改进了的提升管或流化床反应器。本发明提供的方法既可以在单独的提升管或流化床反应器中进行，也可以在与另一个提升管或流化床反应器联合的反应系统中实施。

下面以五种实施方式来具体说明本发明提供的方法，但本发明提供的方法并不局限于上述五种实施方式。

实施方式之一：对于现有提升管反应器的催化裂化装置，将常规的裂化原料更换为汽油原料，需要在再生器之后增加一个冷却器，或者将再生器和冷却器连成为上下一体，将再生催化剂冷却至 100~600℃，然后与汽油原料接触，生成的油气进入沉降器与待生催化剂分离后进入分馏系统。

实施方式之二：对于单提升管反应器的催化裂化装置，需要新建一个提升管反应器和一个冷却器。新建反应器与原有反应器共用沉降器、汽提器、分馏系统和再生器。新建反应器的原料为汽油原料，该反应器称为汽油提升管；原有反应器的原料为常规的裂化原料，该反应器称为原料油提升管。汽油原料和常规的裂化原料分别在汽油提升管和原料油提升管中反应，生成的

混合及回原料油提升管，一部分经冷却器冷却后返回汽油提升管

2

实施方式之三：对于单提升管反应器的催化裂化装置，需要新建一个冷却器和一个带或不带提升管的流化床反应器，该反应器带或不带汽提段。新建反应器与原有反应器共用再生器。新建反应器的原料为汽油原料，该反应器称为汽油反应器；原有反应器的原料为常规的裂化原料，该反应器称为原料油提升管。汽油原料和常规的裂化原料分别在汽油反应器和原料油提升管中反应，汽油原料生成的油气和常规的裂化原料生成的油气混合进入催化裂化主分馏塔，或者分别进入各自的分馏系统。待生催化剂经汽提后再生，再生后的催化剂分为两部分，其中一部分返回原料油提升管，另一部分经冷却器返回汽油反应器。

实施方式之四：本实施方式的装置与实施方式之二的相同。切割后的轻汽油馏分（沸点范围为 40~100℃）和重汽油馏分（沸点范围为 100~200℃）分别从汽油提升管的底部和中上部进入汽油提升管，与催化剂接触反应；同时预热后的常规裂化原料从原料油提升管底部进入提升管，与热的再生催化剂接触。生成的油气均进入沉降器、分馏系统，待生催化剂经汽提后再生，再生后的催化剂分为两部分，其中一部分返回原料油提升管，另一部分经冷却器冷却后返回汽油提升管。

实施方式之五：本发明提供的方法还可以在与一种新型提升管反应器联合的反应系统中实施。需要新建一个新型提升管反应器和一个冷却器。将原有提升管中的常规裂化原料更换为汽油原料，该反应器称为汽油提升管；新建的新型提升管反应器的原料为常规的裂化原料，该反应器称为原料油提升管。汽油在汽油提升管中进行反应，生成的油气进入原料油提升管的第二反应区作为冷激介质，原料油提升管中的油气和待生催化剂通过其出口区进入沉降器。油气进入分离系统分离，待生催化剂经汽提、再生后分为两部分，其中一部分返回新型提升管，另一部分经冷却后返回汽油提升管。

该新型提升管反应器如申请号为 99105903.4 的发明专利申请中所述，沿垂直方向从下至上依次为互为同轴的预提升段、第一反应区、直径扩大了的第二反应区、直径缩小了的出口区，在出口区末端连有一段水平管。第一、二反应区的结合部位为圆台形，其纵剖面等腰梯形的顶角 α 为 30~80°；第二反应区与出口区的结合部位为圆台形，其纵剖面等腰梯形的底角 β 为 45~85°。

一种实施方式的描述，设备和管线的形状和尺寸不受附图的限制，可以根据

3

具体情况确定.

如图1所示，第一种实施方式的流程如下：

预热后的汽油原料经管线 1 进入提升管 2 底部，与来自再生斜管 17 的再生催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器 7，油气和水蒸汽经管线 8 进入分离系统 9，气体和汽油产品经管线 10 出装置，而柴油产品则经管线 11 出装置。待生催化剂进入汽提器 3，由来自管线 4 的水蒸汽汽提后，由待生斜管 5 进入再生器 13。待生催化剂在空气中烧焦再生，空气经管线 14 进入再生器 13、烟气经管线 12 出再生器，热的再生催化剂经管线 15 进入冷却器 16，冷却后由再生斜管 17 返回提升管底部循环使用，松动风经管线 18 进入冷却器。

如图2所示，第二种实施方式的流程如下：

预热后的汽油原料经管线 1 进入提升管 2 底部，与来自再生斜管 17 的再生催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器 27。

同时预提升介质经管线 20 从原料油提升管 22 的底部进入，热的再生催化剂经再生斜管 19 进入提升管 22 的底部由预提升介质进行提升，预热后的常规裂化原料经管线 21 进入提升管 22 的底部，与热催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器 27。

油气和水蒸汽经管线 28 进入分离系统 29，柴油和重油经管线 30 出装置，气体和汽油产品经管线 31 出装置。

待生催化剂进入汽提器 23，由来自管线 24 的水蒸汽汽提后，由待生斜管 25 进入再生器 13。待生催化剂在空气中烧焦再生，空气经管线 14 进入再生器 13，烟气经管线 12 出再生器，热的再生催化剂分为两部分，其中一部分经再生斜管 19 返回原料油提升管 22，另一部分经管线 15 进入冷却器 16，冷却后由再生斜管 17 返回汽油提升管 2 循环使用，松动风经管线 18 进入冷却器。

如图3所示，第三种实施方式的流程如下：

预热后的汽油原料经管线 1 进入汽油提升管 2 底部，与来自再生斜管 17 的再生催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器 7，油气和水蒸汽经管线 8 进入分离系统 9，气体和汽油产品经

4

3、23通过管线33相连，可以只共用汽提器23。

同时预提升介质经管线20从原料油提升管22的底部进入，热的再生催化剂经再生斜管19进入提升管22的底部由预提升介质进行提升，预热后的常规裂化原料经管线21进入提升管22的底部，与热催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器27，油气和水蒸汽或来自管线32的油气一起经管线28进入分离系统29，气体和汽油产品经管线31出装置，而柴油和重油则经管线30出装置。待生催化剂进入汽提器23，由来自管线24的水蒸汽汽提后，由待生斜管25进入再生器13。

待生催化剂在再生器13中烧焦再生，空气经管线14进入再生器，烟气经管线12流出，热的再生催化剂分为两部分，其中一部分经再生斜管19进入原料油提升管22，另一部分经管线15进入冷却器16，冷却后由再生斜管17返回汽油提升管2循环使用，松动风经管线18进入冷却器。

如图4所示，第四种实施方式的流程如下：

预热后的轻汽油馏分（沸点范围为40~100℃）经管线1进入汽油提升管2底部，与来自再生斜管17的再生催化剂混合后进行反应，重汽油馏分（沸点范围为100~200℃）经管线34进入提升管2的中上部继续反应，反应物流进入带有或不带有密相流化床反应器的沉降器27。

同时预提升介质经管线20从原料油提升管22的底部进入，热的再生催化剂经再生斜管19进入提升管22的底部由预提升介质进行提升，预热后的常规裂化原料经管线21进入提升管22的底部，与热催化剂混合后进行反应，反应物流进入带有或不带有密相流化床反应器的沉降器27。油气和水蒸汽经管线28进入分离系统29，柴油和重油经管线30出装置，气体和汽油产品经管线31出装置。

待生催化剂进入汽提器23，由来自管线24的水蒸汽汽提后，由待生斜管25进入再生器13。待生催化剂在空气中烧焦再生，空气经管线14进入再生器13，烟气经管线12出再生器，热的再生催化剂分为两部分，其中一部分经再生斜管19返回提升管22，另一部分由管线15经冷却器16，通过再生斜管17返回汽油提升管2循环使用，松动风经管线18进入冷却器。

如图5所示，第五种实施方式的流程如下：

预热后的汽油原料经管线1进入汽油提升管2的底部，冷却后的再生催

5

剂经再生斜管 19 进入提升管 22 的预提升段 a 由预提升介质进行提升，预热后的常规裂化原料经管线 21 进入提升管 22，与热催化剂混合后在提升管 22 的第一反应区 b 进行反应，反应物流进入提升管 22 的第二反应区 c，与来自汽油提升管 2 的反应物流一起进行二次反应。

油气和待生催化剂经提升管 22 的出口区 d、水平管 e 进入沉降器 27，油气和水蒸汽经管线 28 进入分离系统 29，气体和汽油产品经管线 31 出装置，柴油和重油则经管线 30 出装置。

待生催化剂进入汽提器 23，由来自管线 24 的水蒸汽汽提后，由待生斜管 25 进入再生器 13，待生催化剂在再生器中烧焦再生，空气经管线 14 进入再生器，烟气经管线 12 流出，热的再生催化剂分为两部分，其中一部分经再生斜管 19 进入提升管 22，另一部分经管线 15 进入冷却器 16，冷却后由再生斜管 17 返回汽油提升管 2 循环使用，松动风经管线 18 进入冷却器。

本发明的优点在于：

1、本发明采用提升管或流化床反应器进行连续反应一再生循环操作方式进行催化转化降低汽油烯烃和硫、氮含量；

2、本发明可以处理烯烃含量为 20~90 重%的汽油，适用不同类型催化剂；

3、该方法可以将劣质汽油原料的硫含量降低 1/4~3/4，氮含量可以降至 1ppm 左右。

4、本发明可以与现有的催化裂化反应装置共用分馏系统、汽提器和再生器，只要再增加一个汽油原料的提升管或流化床反应器和再生剂冷却器。

图 1~5 分别为降低汽油烯烃含量的方法的第一至五种实施方式的流程示意图，图中各编号说明如下：1、4、8、10、11、12、14、15、18、20、21、24、28、30、31、32、33、34 均代表管线；2 为汽油提升管反应器，3、23 为汽提器，5、25 为待生斜管，6、26 为分布板、7、27 为沉降器（带或不带流化床反应器），9、29 为分离系统，13 为再生器，16 为冷却器，17、19 为再生斜管，22 为原料油提升管反应器。图 6 为新型提升管反应器的示意图，图中的 a、b、c、d、e 分别代表预提升段、第一反应区、第二反应区、出口区、水平管。

下面的实施例将对本发明予以进一步说明，但并不因此而限制本发明。

实施例 1

本实施例说明采用本发明提供的方法，使用不同类型的催化剂在小型流化床反应器内催化转化降低汽油烯烃的情况.

以表 2 所列的汽油 A 为原料，使用表 1 所列四种不同类型的催化剂，在连续反应再生操作的小型流化床反应器内进行汽油催化转化降低烯烃试验。汽油原料与高温水蒸汽混合后进入流化床反应器内，在反应温度为 300℃，反应器顶部压力为 0.2 兆帕，进料重时空速为 4 小时$^{-1}$，剂油比为 6:1，水油比为 0.03:1 的条件下与催化剂接触进行催化转化反应。反应产物、蒸汽和待生催化剂在沉降器内分离，分离反应产物得到气体产物和液体产物，而待生催化剂进入汽提器，由水蒸汽汽提出待生催化剂上吸附的烃类产物。汽提后的催化剂进入到再生器，与加热过的热空气接触进行再生，再生后的催化剂冷却再返回到反应器循环使用。试验条件、试验结果和汽油的性质列于表 3.

从表 3 可以看出，催化剂类型不同对汽油原料催化转化影响程度也不同。汽油组成中的异构烷烃占 42~55 重 %、芳烃占 25.0~27.0 重 %、烯烃仅占 8.5~18.5 重 %，汽油中的硫含量降到 40~125ppm，氮含量降到 0.4~0.85ppm.

实施例 2

本实施例说明采用本发明提供的方法，使用不同烯烃含量的汽油在小型流化床反应器内催化转化降低汽油烯烃的情况.

以表 2 所列的四种汽油为原料，使用表 1 所列的催化剂 A，在连续反应再生操作的小型流化床反应器内进行汽油催化转化降低烯烃试验。汽油原料与高温水蒸汽混合后进入流化床反应器内，与催化剂接触进行催化转化反应。反应产物、蒸汽和待生催化剂在沉降器内分离，反应产物去分离得到气体产物和液体产物，而待生催化剂进入汽提器，由水蒸汽汽提出待生催化剂上吸附的烃类产物。汽提后的催化剂进入到再生器，与加热过的热空气接触进行再生，再生后的催化剂冷却再返回到反应器循环使用。

试验条件、试验结果和汽油的性质列于表 4，从表 4 可以看出，大部分烯烃

氮含量降到 0.4~1.6ppm．烯烃含量越高的汽油经催化转化其组成中的异构烷烃含量也越高．

实施例 3

本实施例说明采用本发明提供的方法，汽油原料在不同的操作条件下在小型流化床反应器内催化转化降低汽油烯烃情况。

以表 2 所列的汽油 A 为原料，使用表 1 所列的催化剂 A，在连续反应再生操作的小型流化床反应器内进行汽油催化转化降低烯烃试验。汽油原料与高温水蒸汽混合后进入流化床反应器内，在反应温度为 250～450℃、反应器顶部压力为 0.2 兆帕、进料重时空速为 4~10 小时$^{-1}$、剂油比为 3~8:1、水油比为 0.03~0.05:1 的条件下与催化剂接触进行催化转化反应。反应产物、蒸汽和待生催化剂在沉降器内分离，反应产物去分离得到气体产物和液体产物，而待生催化剂进入汽提器，由水蒸汽汽提出待生催化剂上吸附的烃类产物。汽提后的催化剂进入到再生器，与加热过的热空气接触进行再生，再生后的催化剂冷却再返回到反应器循环使用。试验条件、试验结果和汽油的性质列于表 5.

从表 5 可以看出，不同的操作条件对汽油原料催化转化影响程度不同，汽油组成中的异构烷烃占 52.5~57.0 重 %、芳烃占 25.0~26.5 重 %、烯烃仅占 5.5~9.5 重 %，汽油中的硫含量降到 36~46ppm，氮含量降到 0.3~0.41ppm.

实施例 4

本实施例说明采用本发明提供的方法，富含烯烃的汽油在中型提升管反应器内催化转化降低汽油烯烃的情况。可以用该试验结果模拟双提升管反应器中的汽油提升管。

以表 2 所列的汽油 B 为原料，使用表 1 所列的催化剂 C，在连续反应再生操作的中型提升管反应器内进行汽油催化转化降低烯烃、硫、氮含量的试验。汽油原料与高温水蒸汽混合后进入提升管底部，与 550℃再生催化剂接触进行催化转化反应，反应条件如下：反应温度 550℃，反应器顶部压力为 0.2 兆帕，进料重时空速为 50 小时$^{-1}$，剂油比为 6:1，水油比为 0.03:1．反应产物、蒸汽和待生催化剂在沉降器内分离，反应产物去分离得到气体产物

8

于表6，汽油的性质列于表7.

从表7可以看出，汽油组成中的异构烷烃占 40.32 重%、芳烃占 30.16 重%、烯烃仅占 16.49 重%，汽油中的硫含量降到 97ppm，氮含量降到 0.76ppm，此外液化气产率为 14.60 重%，其中丙烯为 3.83 重%；异丁烷为 5.58 重%，而干气仅为 0.66 重%.

实施例 5

本实施例说明采用本发明提供的方法，将富含烯烃的汽油切割为轻的窄馏分和重的窄馏分，这两种窄馏分分别从中型提升管反应器的底部、中上部进入，进行催化转化降低汽油烯烃、硫、氮的试验.

以表2所列的轻汽油馏分C和重汽油馏分D为原料，使用表1所列的催化剂A，在连续反应再生操作的中型提升管反应器内进行汽油催化转化降低烯烃、硫、氮含量的试验。轻汽油馏分 C 与高温水蒸汽混合后进入提升管底部，与温度为 300℃的再生催化剂接触进行催化转化反应；同时重汽油馏分D进入提升管中部，与温度为 400℃的催化剂接触进行催化转化反应。反应条件为：反应器顶部压力为 0.2 兆帕，进料重时空速为 50~100 小时⁻¹，催化剂与原料油的重量比为 6:1，水蒸汽与原料油的重量比为 0.03:1。反应产物、蒸汽和待生催化剂在沉降器内分离，反应产物去分离得到气体产物和液体产物，而待生催化剂进入汽提器，由水蒸汽汽提出待生催化剂上吸附的烃类产物。汽提后的催化剂进入到再生器，与加热过的热空气接触进行再生，再生后的催化剂冷却再返回到反应器循环使用。试验条件、试验结果列于表6和汽油的性质列于表7.

从表 6 可以看出，气体收率为 6.99 重%,柴油的收率为 4.53 重%，而汽油的收率为 86.78 重%，从表 7 可以看出，汽油组成中的异构烷烃占 51.25 重%、芳烃占 26.98 重%、烯烃仅占 8.59 重%，汽油中的硫含量降到 86ppm，氮含量降到 0.65ppm.

实施例 6

本实施例说明采用本发明提供的方法，在小型固定流化床反应器内催化转化降低汽油烯烃的情况.

以表 2 所列的汽油 以 为原料，使用表 1 所列的催化剂 A，在连续反应再生操作

高温水蒸汽混合后进入 流化床反应器 与 入 温度

9

器顶部压力为 0.2 兆帕、进料重时空速为 4 小时$^{-1}$、剂油比为 6:1，水油比为 0~0.03:1 的条件下与催化剂接触进行催化转化反应。反应产物、蒸汽和待生催化剂在沉降器内分离，反应产物去分离得到气体产物和液体产物，而待生催化剂进入汽提器，由水蒸汽汽提出待生催化剂上吸附的烃类产物。汽提后的催化剂进入到再生器，与加热过的热空气接触进行再生，再生后的催化剂冷却再返回到反应器循环使用。试验条件、试验结果和汽油的性质列于表 8.

· 从表 8 可以看出，汽油组成中的异构烷烃占 61~65 重 %、芳烃占 6.5~7.0 重 %、烯烃仅占 15.5~20.0 重 %，汽油中的硫含量降到 100~140ppm，氮含量降到 0.65~1.10ppm.

表1

汽油原料编号	A	B	C	D
密度(20℃)，千克/米³	743.0	727.1	653.1	786.4
辛烷值				
RON	91.3	92.1	93.2	88.6
MON	79.5	79.8	81.5	78.4
硫，ppm	200	2035.5	376.7	2892.2
氮，ppm	30	151.3	14.3	93.0
碳，重%	86.80	86.54	85.26	86.29
氢，重%	13.03	13.26	14.52	12.99
馏程，℃				
初馏点	49	44	44	90
10%	63	59	48	91
30%	82	78	53	120
50%	106	104	60	153
70%	140	133	65	173
90%	176	166	75	186
终馏点	195	200	90	202
族组成，重%				
烷烃	29.64	26.94	19.04	25.00
正构烷烃	5.29	4.88	4.98	5.12
异构烷烃	25.35	22.06	14.06	19.88
环烷烃	8.25	7.16	5.17	12.30
烯烃	37.79	47.65	69.64	13.62
芳烃	24.32	18.25	6.15	49.08

11

表2

催化剂编号	A	B	C	D
商品牌号	CRC-1	RHZ-200	ZCM-7	RAG-1
沸石类型	REY	REHY	USY	REY-USY-ZRP
化学组成，重%				
氧化铝	26.5	33.0	46.4	44.6
氧化钠	0.19	0.29	0.22	0.13
氧化铁	0.09	1.1	0.32	—
表观密度，千克/米^3	450	560	690	620
孔体积，毫升/克	0.41	0.25	0.38	0.36
比表面积，米^2/克	132	92	164	232
磨损指数，重%时$^{-1}$	4.2	3.2	—	2.5
筛分组成，重%				
0~40 微米	7.3	15.2	4.8	13.1
40~80 微米	43.7	55.1	47.9	54.9
>80 微米	49.0	29.7	47.3	32.0

12

表3

催化剂		A	B	C	D
反应温度，℃		300	300	300	300
重时空速，小时$^{-1}$		4	4	4	4
剂油比		6	6	6	6
水油比		0.03	0.03	0.03	0.03
产品分布，重%					
干气		1.36	0.87	0.65	0.56
液化气		3.87	4.69	4.76	4.93
汽油		85.97	86.85	88.70	89.95
轻柴油		4.37	3.98	3.01	2.36
重柴油		2.43	2.02	1.63	1.23
焦炭		1.98	1.56	1.23	0.93
损失		0.02	0.03	0.02	0.04
汽油性质	原料性质				
RON	91.3	88.2	89.3	90.2	90.5
MON	79.5	80.1	80.0	79.8	79.8
硫，ppm	200	40	65	102	125
氮，ppm	30	0.4	0.7	0.76	0.85
芳烃，重%	24.32	25.20	25.00	25.30	26.56
烯烃，重%	37.79	8.70	10.87	14.98	18.39
烷烃，重%	29.64	59.01	56.92	52.29	47.19
正构烷烃	5.29	5.01	5.05	4.98	4.86
异构烷烃	25.35	54.0	51.87	47.31	42.33
环烷烃,重%	8.25	7.09	7.21	7.43	7.86

13

表4

原料油	A	B	C	D
反应温度，℃	300	300	300	400
重时空速，小时⁻¹	4	4	4	4
剂油比	6	6	6	6
水油比	0.03	0.03	0.03	0.03
产品分布，重%				
干气	1.36	1.57	0.73	1.33
液化气	3.87	4.64	6.05	7.14
汽油	85.97	84.11	86.39	81.83
轻柴油	4.37	5.04	3.33	4.32
重柴油	2.43	2.61	1.36	2.04
焦炭	1.98	2.02	2.12	3.14
损失	0.02	0.01	0.02	0.20
汽油性质				
RON	88.2	90.8	90.2	89.4
MON	80.1	81.0	81.2	78.6
硫，ppm	40	402	108.5	578
氮，ppm	0.4	1.0	0.83	1.6
芳烃，重%	25.20	19.20	6.52	55.18
烯烃，重%	8.70	10.92	16.34	3.89
烷烃，重%	59.01	62.72	70.02	27.48
正构烷烃	5.01	4.95	5.20	5.22
异构烷烃	54.0	57.77	64.82	22.26
环烷烃，重%	7.09	7.16	7.12	13.45

表 5

操作条件							
反应温度, ℃		300	300	300	300	250	450
重时空速, 小时$^{-1}$		8	4	4	4	4	10
剂油比		3	6	8	6	6	6
水油比		0.03	0.03	0.03	0.05	0.03	0.05
产品分布, 重%							
干气		1.12	1.36	1.56	1.13	0.56	3.41
液化气		3.55	3.87	4.32	3.53	1.87	5.48
汽油		87.11	85.97	84.54	86.61	87.02	82.79
轻柴油		4.01	4.37	4.63	4.18	5.47	3.24
重柴油		2.11	2.43	2.75	2.33	2.86	1.36
焦炭		1.86	1.98	2.02	1.95	1.96	2.93
损失		0.24	0.02	0.18	0.27	0.26	0.29
汽油性质	原料性质						
RON	91.3	89.0	88.2	87.8	89.0	87.6	88.8
MON	79.5	80.0	80.1	80.1	79.9	80.0	80.0
硫, ppm	200	46	40	36	42	36	36
氮, ppm	30	0.41	0.4	0.3	0.4	0.3	0.3
芳烃, 重%	24.32	25.12	25.20	26.45	25.01	25.03	26.02
烯烃, 重%	37.79	9.32	8.70	6.67	9.13	5.98	9.09
烷烃, 重%	29.64	58.45	59.01	59.80	58.76	61.84	57.80
正构烷烃	5.29	5.07	5.01	5.11	5.07	5.03	4.98
异构烷烃	25.35	53.38	54.0	54.69	53.69	56.81	52.82
环烷烃, 重%	8.25	7.11	7.09	7.08	7.10	7.15	7.09

15

表6

	实施例4	实施例5
操作条件		
反应温度，℃	550	300/400*
重时空速，小时⁻¹	50	50/100*
剂油比	6.0	6.0
水油比	0.03	0.03
产品分布，重%		
干气	0.66	1.01
液化气	14.60	5.98
其中丙烯	3.83	
异丁烷	5.58	
汽油	75.62	86.78
轻柴油	3.75	3.52
重柴油	2.53	1.01
焦炭	2.43	1.65
损失	0.41	0.05

*："/"之前的数值表示汽油提升管底部的反应条件，"/"之后的数值表示汽油提升管中部的反应条件。

16

表7

	原料 A	实施例 4	原料 C	原料 D	实施例 5
密度(20℃)，千克/米³	743.0	741.8	653.1	786.4	746.7
辛烷值					
RON	91.3	90.0	93.2	88.6	91.0
MON	79.5	80.8	81.5	78.4	81.2
硫，ppm	200	97	376.7	2892.2	86
氮，ppm	30	0.76	14.3	93.0	0.65
碳，重%	86.80	86.71	85.26	86.29	86.43
氢，重%	13.03	13.22	14.52	12.99	13.42
馏程，℃					
初馏点	49	49	44	90	46
10%	63	75	48	91	72
30%	82	98	53	120	90
50%	106	120	60	153	118
70%	140	147	65	173	143
90%	176	178	75	186	175
终馏点	195	202	90	202	196
族组成，重%					
芳烃	24.32	30.86	6.15	49.08	26.98
烯烃	37.79	16.49	69.64	13.62	8.59
烷烃	29.64	45.34	19.04	25.00	56.57
正构烷烃	5.29	5.02	4.98	5.12	5.32
异构烷烃	25.35	40.32	14.06	19.88	51.25
环烷烃	8.25	7.31	5.17	12.30	7.86

17

表 8

操作条件		150	200	250	300
反应温度，℃		150	200	250	300
重时空速，小时⁻¹		4	4	4	4
剂油比		6	6	6	6
水油比		0	0.02	0.02	0.03
产品分布，重%					
干气		0.86	0.51	0.64	0.73
液化气		2.04	2.68	4.30	6.05
汽油		93.24	93.23	90.02	86.39
轻柴油		1.65	1.52	2.46	3.33
重柴油		0.76	0.73	1.09	1.36
焦炭		1.33	1.30	1.40	2.12
损失		0.12	0.03	0.09	0.02
汽油性质	原料性质				
RON	93.2	88.2	89.8	90.0	90.2
MON	81.5	81.2	81.5	81.5	81.5
硫，ppm	376.7	106.5	137.2	101.3	108.5
氮，ppm	14.3	0.65	1.10	0.65	0.83
芳烃，重%	6.15	6.55	6.66	6.50	6.52
烯烃，重%	69.64	16.70	19.53	15.98	16.34
烷烃，重%	19.04	68.74	66.49	70.30	70.02
正构烷烃	4.98	5.30	5.33	5.17	5.20
异构烷烃	14.06	63.44	61.16	65.13	64.82
环烷烃，重%	5.17	8.01	7.32	7.22	7.12

说　明　书　附　图

图1

图2

Fig. 3

图4

图5

去分离系统

待生催化剂
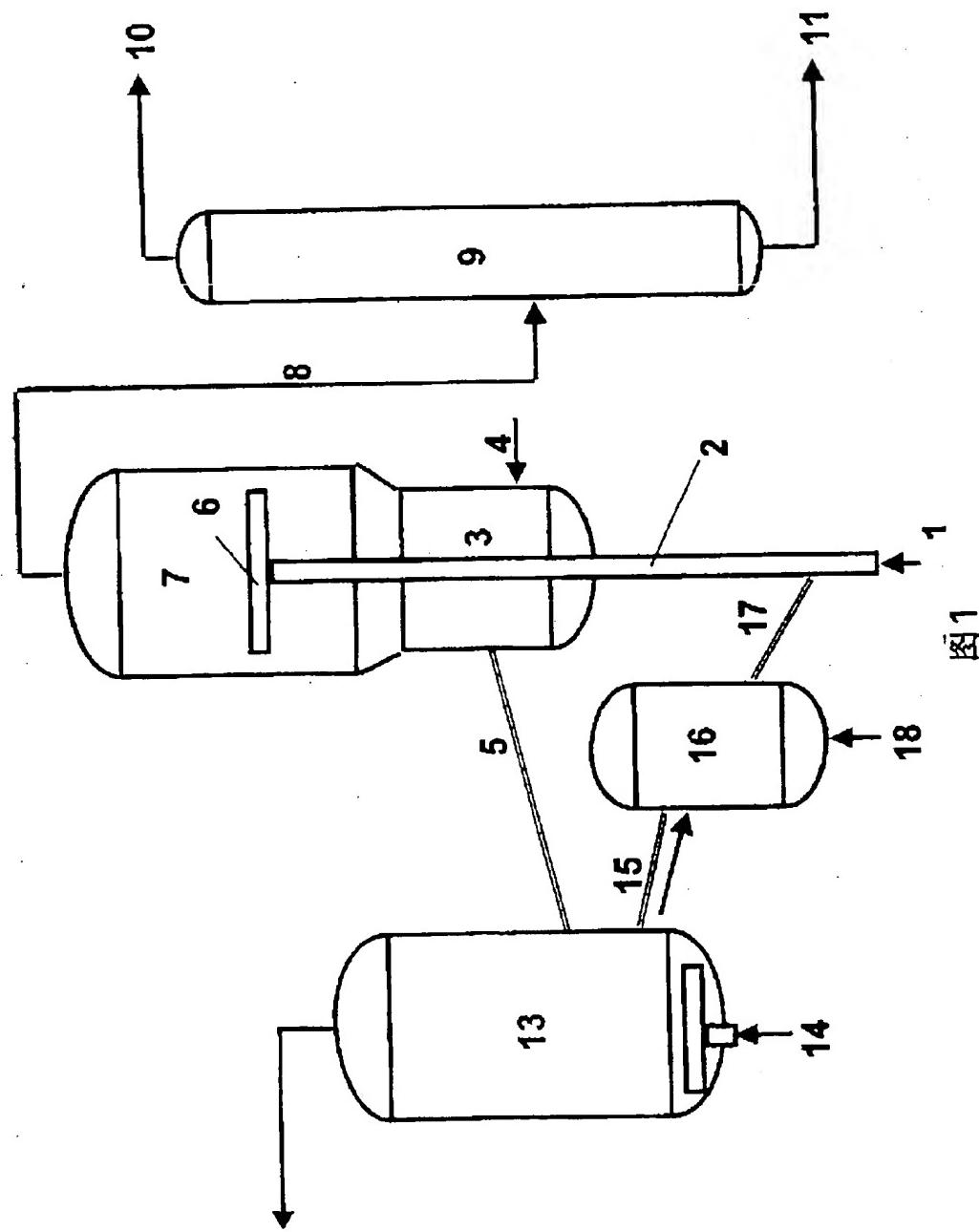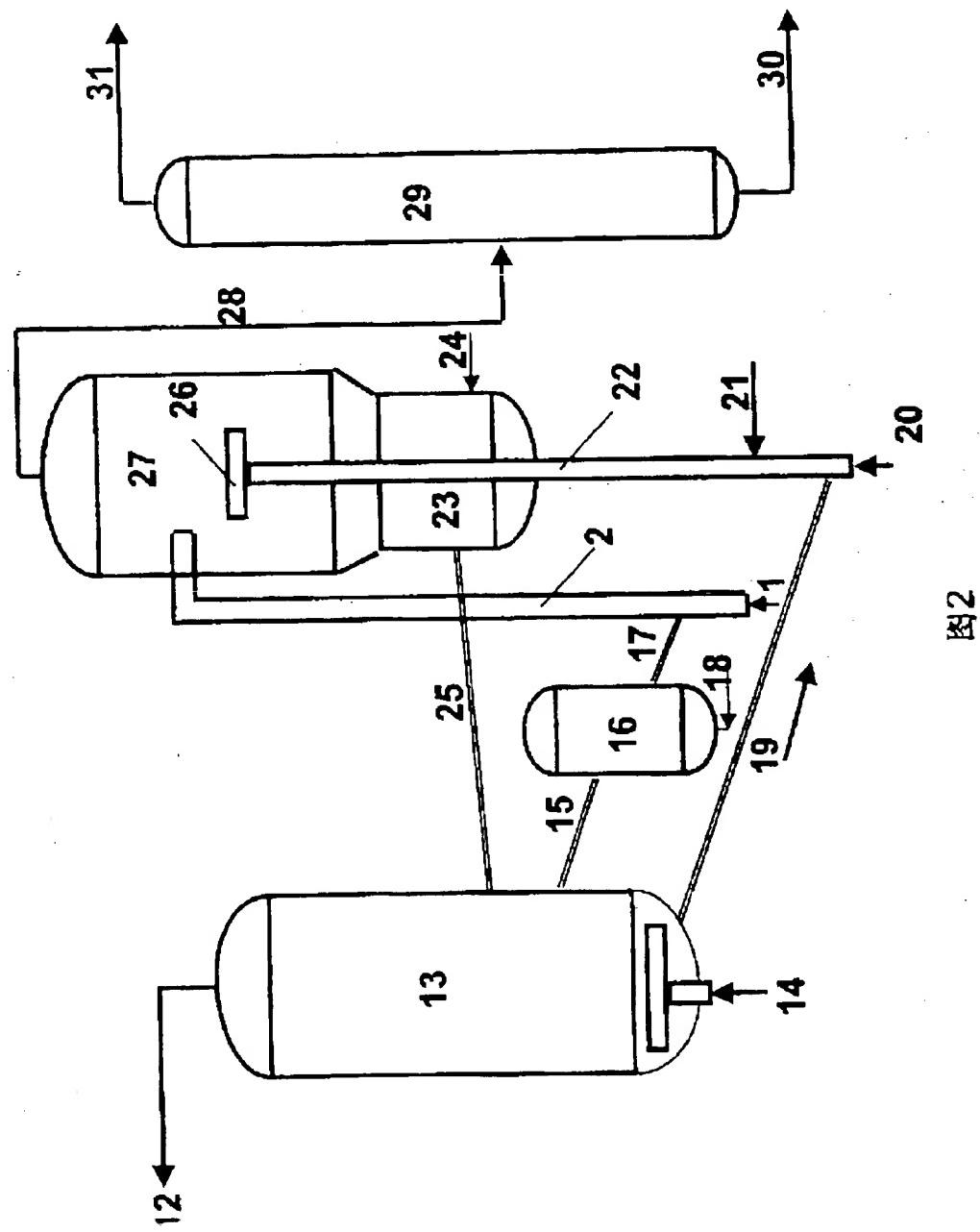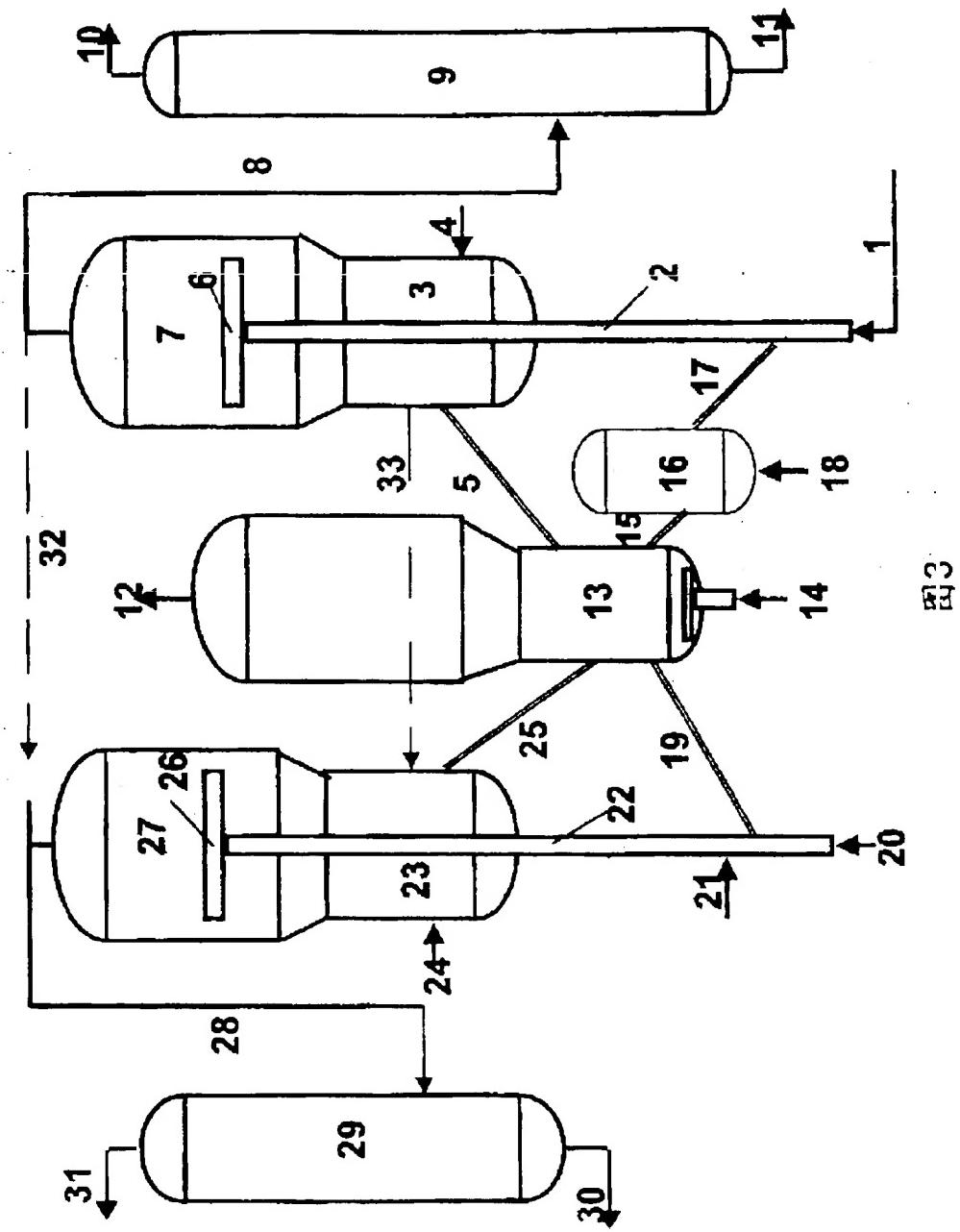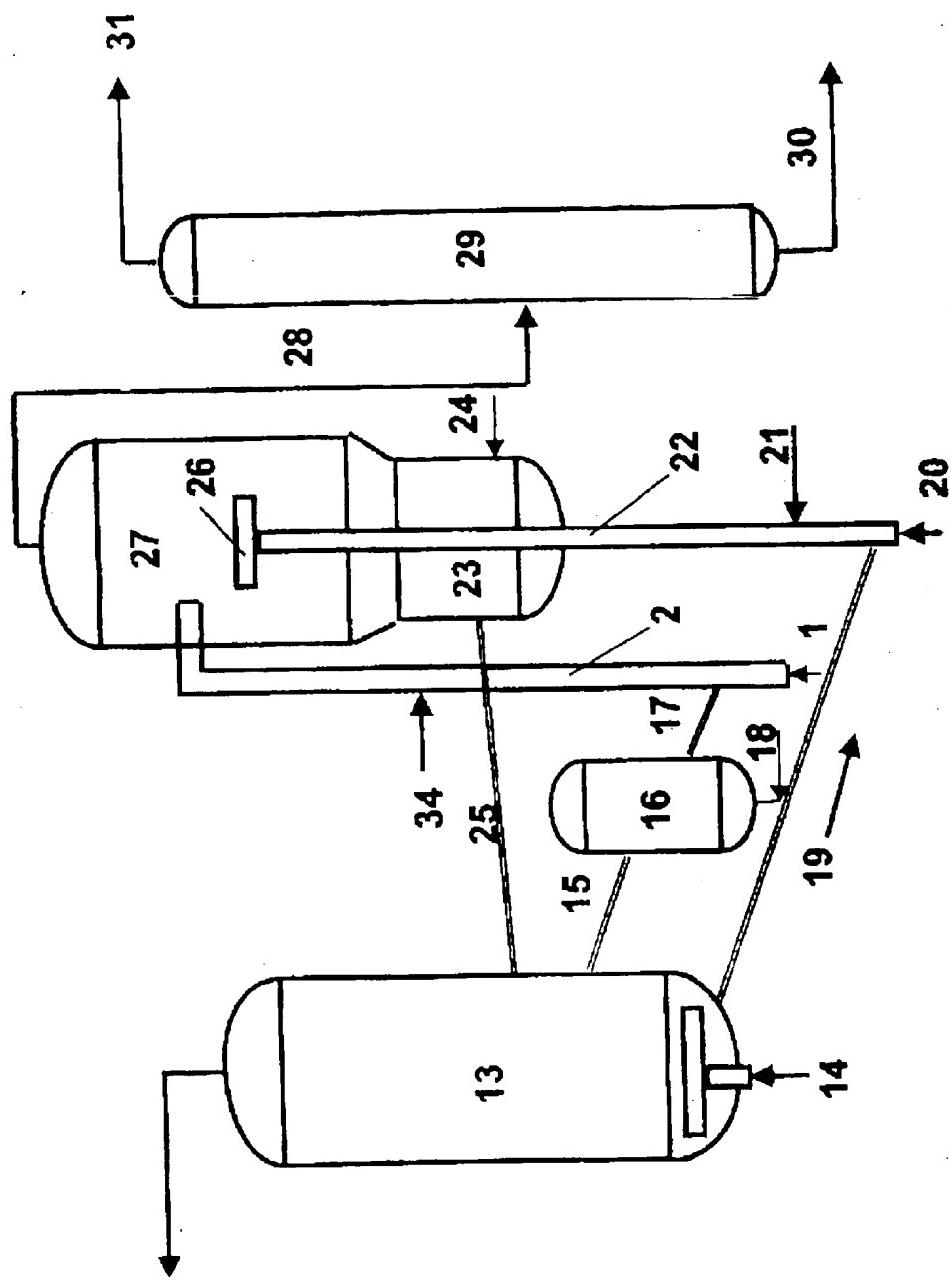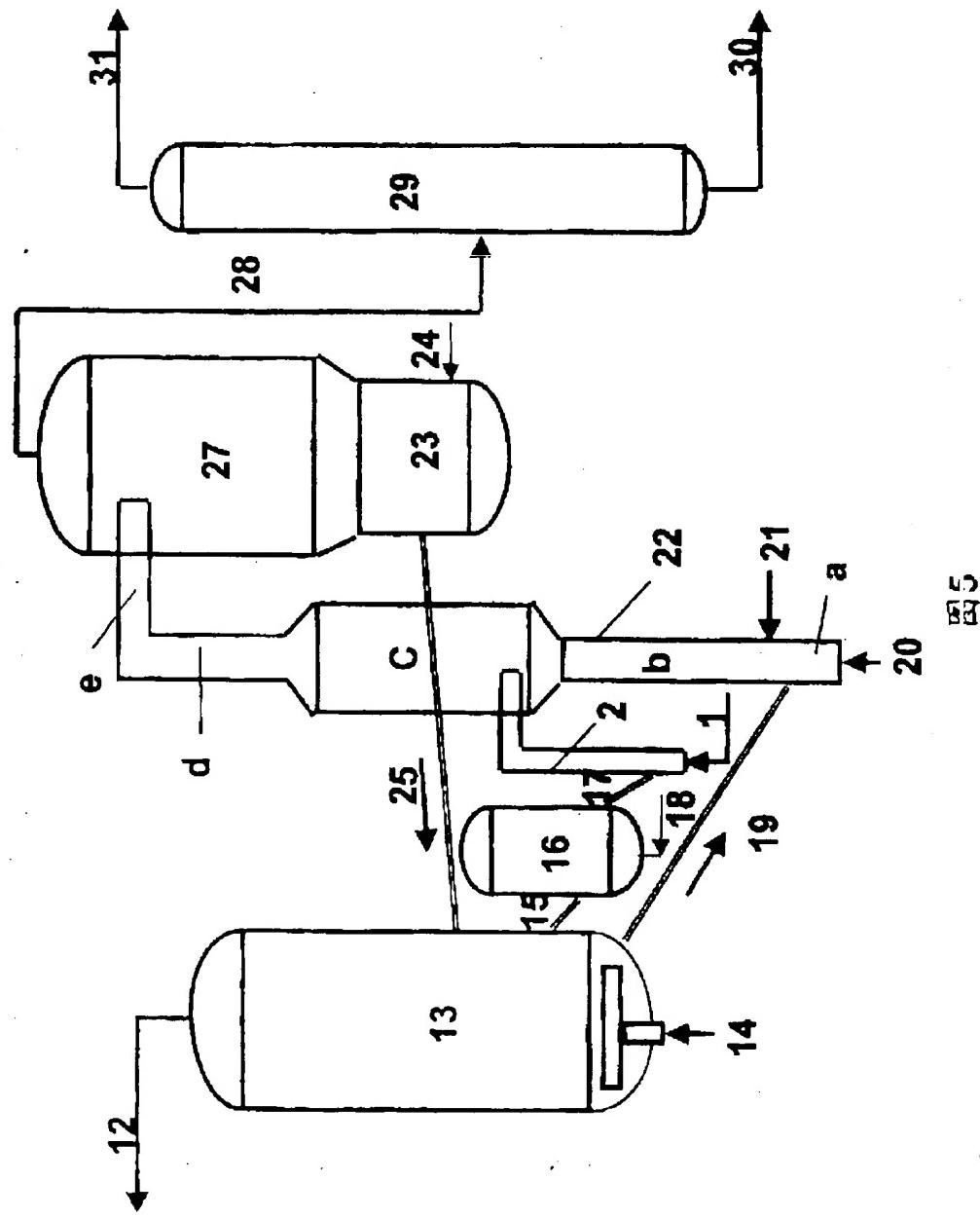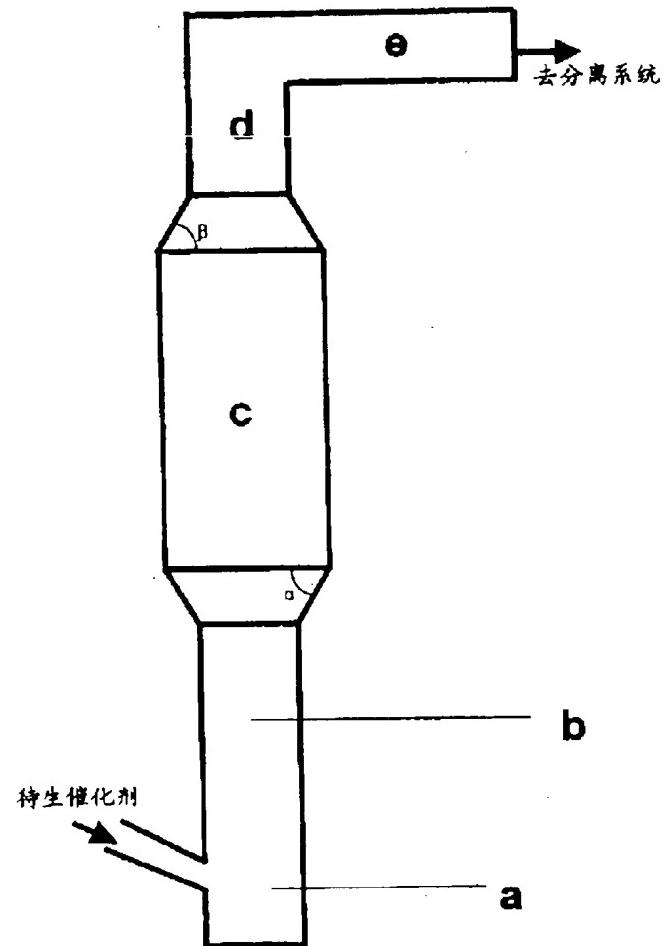
图6

TRANSLATION FROM CHINESE

[19] INTELLECTUAL PROPERTIES OFFICE, PEOPLE'S REPUBLIC OF CHINA

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[51] Int.Cl.^{7}

C10G 35/00

C10G 45/02

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[74] Patent Agency: Patent Office, SINOPEC Petrochemical Research Institute

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Number of Description Pages: 18

Sulfur, and Nitrogen Contents in Gasoline

[57]   Abstract:

This Invention relates to a catalytic conversion process for reducing the olefin, sulfur, and nitrogen contents in gasoline, wherein pre-heated crude gasoline enters a riser or fluid bed reactor and reacts with the catalyst upon contact under the following reaction conditions: the temperature at 100-600°C, the weight-space velocity at 1-120 hours^{-1}, the weight ratio of the catalyst and the crude gasoline at 2-15:1, and the weight ratio of the steam and the crude gasoline at 0-0.10:1. The products of the said catalytic conversion are separated, and the regenerable catalyst is recycled, through stripping, regenerating, and cooling, back to the reactor for re-use. The said process of this Invention is capable of reducing the olefin content in gasoline to 20wt% and increasing the isomerized alkane content in gasoline to 66wt%.

CLAIMS

1.   A catalytic conversion process for reducing the olefin, sulfur, and nitrogen contents in gasoline, wherein pre-heated crude gasoline enters a riser or fluid bed reactor and reacts with the catalyst upon contact under the following reaction conditions: the temperature at 100-600°C, the weight-space velocity at 1-120 hours^{-1}, the weight ratio of the catalyst and the crude gasoline at 2-15:1, and the weight ratio of the steam and the crude gasoline at 0-0.10:1. The products of the said catalytic conversion are separated, and the regenerable catalyst is recycled, through stripping, regenerating, and cooling, back to the reactor for re-use.

2.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the said process can be implemented either in a single riser or fluid bed reactor or in a unit of two coupled riser or fluid bed reactors.

3.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the cooling of the regenerable catalyst is performed in a cooler, which may be coupled with a regenerator to form a combined unit or be connected to the regenerator through piping.

4.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the active ingredients of the said catalyst are taken from Y or HY zeolite with or without rare earth elements, super stable Y zeolite with or without rare earth elements, ZSM-5 series zeolite, or one, two, or three kinds of high silicon zeolite, β zeolite, or ferrierite of the five-member ring structure.

5.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the said catalyst may be an amorphous silica-alumina catalyst.

6.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the olefin content in the said crude gasoline is 20-90wt%.

7.   The catalytic conversion process as described in the first paragraph of CLAIMS, wherein the reaction temperature is 150-550°C, the feeding weight-space velocity is 2-

DESCRIPTION

A Catalytic Conversion Process for Reducing the Olefin, Sulfur, and Nitrogen Contents in Gasoline

This Invention relates to a catalytic process for converting petroleum hydrocarbons, and more specifically, to a catalytic conversion process for reducing the olefin, sulfur, and nitrogen contents in gasoline.

As environmental awareness places increasingly demanding requirements on the quality of gasoline, a transition from the lead-free gasoline to the New Composition gasoline is well under way. The New Composition gasoline has restrictive standards on the vapor pressure and the benzene content. It will eventually have restrictions on the arene and olefin contents as well. The New Composition gasoline standards vary with countries and regions, but have a common general principle, which limits the olefin content in gasoline to less than 20wt% and the sulfur content to less than 100ppm.

Currently, commercial gasoline contains 35-65wt% olefin. Gasoline with a high percentage of olefin has poor thermal stability. On a moving vehicle, the olefin in the gasoline will, under the temperature inside the fuel system of the internal combustion engine, get oxidized and then precipitate onto the inner wall of the main inlet of the carburetor, the inner wall of the idling inlet, and the surface of the throttle. As a result, the vehicle soon begins to demonstrate problems such as irregular idling speeds, retarded fuel supply at acceleration, and insufficient burning of gasoline, which leads to an increased emission of hydrocarbons and carbon monoxide, thus polluting the environment. In addition, diffused olefin vapor reacts with the atmospheric ozone and forms toxic compounds.

On the other hand, as crude oil becomes increasingly heavy, the sulfur content in commercial gasoline rises to approximately 300ppm or over. Such gasoline must be hydrogenated to meet the quality standards of the New Composition. However, that

The purpose of this Invention is to provide a catalytic conversion process for converting the olefin in gasoline to isomerized alkane and arene as well as reducing the sulfur and nitrogen contents in gasoline.

This Invention is implemented in the following manner: pre-heated crude gasoline enters a riser or fluid bed reactor and reacts with the catalyst with or without the presence of steam; the reaction products, the steam (if present), and the regenerable catalyst with the post-reaction coke are separated; the separation of the reaction products results in gasoline products as well as a small amount of dry gas, liquefied gas, and diesel oil; the regenerable catalyst is steam stripped and sent to a regenerator to be regenerated through a coke-burning process in the presence of an oxygenated gas; and, finally, the hot regenerated catalyst is cooled and recycled to the reactor for re-use.

More specifically, this Invention is implemented through the following steps: pre-heated crude gasoline enters a riser or fluid bed reactor and have isomerization and hydrogen transfer reactions with the catalyst upon contact under the following reaction conditions: the temperature at 100-600°C (preferably 150-550°C), the reaction pressure at 130-450kPa (preferably 250-400kPa), the weight-space velocity at 1-120 hours^{-1} (preferably 2-100 hours^{-1}), the weight ratio of the catalyst and the crude gasoline (hereafter referred to as "catalyst/gasoline ratio") at 2-15:1 (preferably 3-10:1), and the weight ratio of the steam and the crude gasoline (hereafter referred to as "water/gasoline ratio") at 0-0.10:1 (preferably 0.01-0.05:1); the reaction products, the steam, and the regenerable catalyst with the post-reaction coke are separated; the separation of the reaction products results in dry gas, propylene- and isobutane-rich liquefied gas as well as isomerized alkane-rich and arene-rich gasoline and diesel oil; the regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst; the stripped catalyst is then sent to a regenerator to be regenerated through a coke-burning process in the presence of an oxygenated gas; and, finally, the hot regenerated catalyst is cooled in a cooler and recycled to the reactor for re-use.

The crude gasoline used in this Invention contains 20-90wt% olefin and a sulfur

The products of this Invention include gasoline and a small amount of gas and diesel oil. The gasoline contains low levels of olefin, sulfur, and nitrogen and relatively high levels of isomerized alkane and arene. The liquefied gas contains high levels of propylene and isobutane.

This Invention may use catalysts with active ingredients taken from Y or HY zeolite with or without rare earth elements, super stable Y zeolite with or without rare earth elements, ZSM-5 series zeolite, or one, two, or three kinds of high silicon zeolite, β zeolite, or ferrierite of the five-member ring structure. It may also use amorphous silica-alumina catalysts.

This Invention may use either a conventional riser or fluid bed reactor or a modified model of the conventional riser or fluid bed reactor. The process of this Invention may be implemented either in a single riser or fluid bed reactor or in a unit of two coupled riser or fluid bed reactors.

The following five application examples offer detailed descriptions of the process as listed in this Invention. However, the said process has many ways of implementation, which include, but are not limited to, those five examples.

Application Example 1: Keep the catalytic cracking unit of the existing riser reactor, but use the gasoline stock in place of the cracking stock. It is necessary to add a cooler after the regenerator, or to couple the regenerator and the cooler as one unit. Cool the regenerated catalyst down to 100-600°C, and then let it react with the gasoline stock. Send the resultant oil vapor through a precipitator, have it separated from the regenerable catalyst, and then send it into the fractional system.

Application Example 2: Keep the catalytic cracking unit of the existing single riser reactor, but install a new riser reactor and a new cooler. The new reactor and the existing reactor share the precipitator, the stripper, the fractional system, and the regenerator. The new reactor, which uses the gasoline stock, is designated as the gasoline riser. The existing reactor, which uses the conventional cracking stock, is designated as the stock oil riser. The gasoline stock undergoes the reaction in the gasoline riser while

to the gasoline riser. The regenerable catalyst is stripped and then regenerated. The regenerated catalyst is divided into two portions: one to go back to the stock oil riser and the other to go back, after being cooled by the cooler, to the gasoline riser.

Application Example 3: Keep the catalytic cracking unit of the existing single riser reactor, but install a new cooler and a new fluid bed reactor with or without a riser. The new reactor may or may not have a stripping section. The new reactor and the existing reactor share the regenerator. The new reactor, which uses the gasoline stock, is designated as the gasoline reactor. The existing reactor, which uses the conventional cracking stock, is designated as the stock oil riser. The gasoline stock undergoes the reaction in the gasoline reactor while the conventional cracking stock undergoes the reaction in the stock oil riser. The resultant oil vapor from the gasoline reactor and the resultant oil vapor from the stock oil riser are mixed and sent into the main catalytic cracking fractional column, or they may go separately into their own fractional systems. The regenerable catalyst is stripped and then regenerated. The regenerated catalyst is divided into two portions: one to go back to the stock oil riser and the other to go through the cooler and then back to the gasoline reactor.

Application Example 4: This application example employs the same kind of equipment as Application Example 2 does. The lighter cut gasoline fractions (boiling point at 40-100°C) and the heavier cut gasoline fractions (boiling point at 100-200°C) enter the gasoline riser at the bottom and the upper section respectively. They react with the catalyst upon contact. Meanwhile, the pre-heated conventional cracking stock enters the stock oil riser at the bottom and reacts with the hot regenerated catalyst. The resultant oil vapor from both risers goes into the precipitator and then into the fractional system. The regenerable catalyst is stripped and then regenerated. The regenerated catalyst is divided into two portions: one to go back to the stock oil riser and the other to go back, after being cooled by the cooler, to the gasoline riser.

Application Example 5: The process as listed in this Invention may also be employed in a reaction system coupled with a new-type riser reactor. It is necessary to

designated as the stock oil riser. The gasoline undergoes the reaction in the gasoline riser, and the resultant oil vapor enters the second reaction section of the stock oil riser to serve as a cold stimulating medium. The oil vapor and the regenerable catalyst in the stock oil riser leave the riser at its outlet section and go into the precipitator. The oil vapor is separated from the catalyst by the separation device. The regenerable catalyst is stripped and then regenerated. The regenerated catalyst is divided into two portions: one to go back to the new-type riser and the other to go back, after being cooled by the cooler, to the gasoline riser.

The said new-type riser reactor is described in the Invention Patent Application No. 99105903.4, wherein the coaxial components are vertically aligned in the following order (from the bottom up): the pre-rising block, the first reaction section, the second reaction section (with an enlarged diameter), and the outlet section (with a reduced diameter). A horizontal pipe is attached to the end of the outlet section. The connection between the first reaction section and the second reaction section is shaped like a round platform, whose longitudinal section is an isosceles trapezoid with the vertex angles (the α angles) at 30-80°. The connection between the second reaction section and the outlet section is shaped like a round platform, whose longitudinal section is an isosceles trapezoid with the base angles (the β angles) at 45-85°.

The following paragraphs use the attached illustrations to provide further descriptions of the process as listed in this Invention.

Figures 1-5 illustrate, respectively, the process flow of the catalytic conversion as represented in Application Examples 1-5 for reducing the olefin, sulfur, and nitrogen contents in gasoline. The equipment and piping forms and dimensions are not accurately represented in the illustrations. They should be determined in light of the actual scenarios.

As is shown in Figure 1, the process flow of Application Example 1 is as follows:

The pre-heated gasoline stock flows through a pipe (1) into the bottom section of the riser (2), where it meets and reacts with the regenerated catalyst coming from the

products flow out of the device by a pipe (10).. The diesel products exit the device by another pipe (11). The regenerable catalyst enters the stripper (3) and, after being stripped by the steam coming from a pipe (4), flows through another pipe (5) into the regenerator (13). The regenerable catalyst is regenerated through the coke-burning process in the presence of air. The air comes into the regenerator (13) through a pipe (14). The fume exits the regenerator through another pipe (12). The hot regenerated catalyst enters the cooler (16) through a pipe (15) and, after being cooled, flows through the regeneration pipe (17) back into the bottom section of the riser for re-use. The disengaging air flow comes into the cooler through a pipe (18).

As is shown in Figure 2, the process flow of Application Example 2 is as follows:

The pre-heated gasoline stock flows through a pipe (1) into the bottom section of the riser (2), where it meets and reacts with the regenerated catalyst coming from the regeneration pipe (17). The reaction products flow into the precipitator (27), which may or may not be equipped with a dense-phase fluid bed reactor.

Meanwhile, the pre-rising medium flows through a pipe (20) into the bottom section of the stock oil riser (22). The hot regenerated catalyst flows through the regeneration pipe (19) into the bottom section of the riser (22), where it is subjected to the rising process by the pre-rising medium. The pre-heated conventional cracking stock flows through a pipe (21) into the bottom section of the riser (22), where it meets and reacts with the hot catalyst. The reaction products flow into the precipitator (27), which may or may not be equipped with a dense-phase fluid bed reactor.

The oil vapor and the steam flow through a pipe (28) into the separation system (29). The diesel and heavier oil products exit the device by a pipe (30). The gases and the gasoline products flow out of the device by another pipe (31).

The regenerable catalyst enters the stripper (23) and, after being stripped by the steam coming from a pipe (24), flows through another pipe (25) into the regenerator (13). The regenerable catalyst is regenerated through the coke-burning process in the presence of air. The air comes into the regenerator (13) through a pipe (14). The fume exits the

cooled, flows through the regeneration pipe (17) back into the gasoline riser (2) for re-use. The disengaging air flow comes into the cooler through a pipe (18).

As is shown in Figure 3, the process flow of Application Example 3 is as follows

The pre-heated gasoline stock flows through a pipe (1) into the bottom section of the riser (2), where it meets and reacts with the regenerated catalyst coming from the regeneration pipe (17). The reaction products flow into the precipitator (7), which may or may not be equipped with a dense-phase fluid bed reactor. The oil vapor and the steam flow through a pipe (8) into the separation system (9). The gases and the gasoline products flow out of the device by a pipe (10). The diesel products exit the device by another pipe (11). Or, the oil vapor and the steam flow sequentially through pipes (32 and 28) into the separation system (29) to be separated. The regenerable catalyst enters the stripper (3) and, after being stripped by the steam coming from a pipe (4), flows through another pipe (5) into the regenerator (13). Or, connect the two strippers (3 and 23) with a pipe (33), and use the stripper (23) alone.

Meanwhile, the pre-rising medium flows through a pipe (20) into the bottom section of the stock oil riser (22). The hot regenerated catalyst flows through the regeneration pipe (19) into the bottom section of the riser (22), where it is subjected to the rising process by the pre-rising medium. The pre-heated conventional cracking stock flows through a pipe (21) into the bottom section of the riser (22), where it meets and reacts with the hot catalyst. The reaction products flow into the precipitator (27), which may or may not be equipped with a dense-phase fluid bed reactor. The oil vapor and the steam (or the oil vapor from another pipe (32)) flow through a pipe (28) into the separation system (29). The gases and the gasoline products flow out of the device by a pipe (31). The diesel and heavier oil products exit the device by another pipe (30). The regenerable catalyst enters the stripper (23) and, after being stripped by the steam coming from a pipe (24), flows through another pipe (25) into the regenerator (13).

The regenerable catalyst is regenerated through the coke-burning process in the regenerator (13). The air comes into the regenerator through a pipe (14). The fume exits

riser (22) while the other portion enters the cooler ... through a pipe ... also the

being cooled, flows through the regeneration pipe (17) back into the gasoline riser (2) for re-use. The disengaging air flow comes into the cooler through a pipe (18).

As is shown in Figure 4, the process flow of Application Example 4 is as follows

The pre-heated lighter gasoline fractions (boiling point at 40-100°C) flow through a pipe (1) into the bottom section of the gasoline riser (2), where they meet and react with the regenerated catalyst coming from the regeneration pipe (17). The heavier gasoline fractions (boiling point at 100-200°C) flow through another pipe (34) into the upper mid section of the riser (2) for further reaction. The reaction products flow into the precipitator (27), which may or may not be equipped with a dense-phase fluid bed reactor.

Meanwhile, the pre-rising medium flows through a pipe (20) into the bottom section of the stock oil riser (22). The hot regenerated catalyst flows through the regeneration pipe (19) into the bottom section of the riser (22), where it is subjected to the rising process by the pre-rising medium. The pre-heated conventional cracking stock flows through a pipe (21) into the bottom section of the riser (22), where it meets and reacts with the hot catalyst. The reaction products flow into the precipitator (27), which may or may not be equipped with a dense-phase fluid bed reactor. The oil vapor and the steam flow through a pipe (28) into the separation system (29). The diesel and heavier oil products exit the device by a pipe (30). The gases and the gasoline products flow out of the device by another pipe (31).

The regenerable catalyst enters the stripper (23) and, after being stripped by the steam coming from a pipe (24), flows through another pipe (25) into the regenerator (13). The regenerable catalyst is regenerated through the coke-burning process in the presence of air. The air comes into the regenerator (13) through a pipe (14). The fume exits the regenerator through another pipe (12). The hot regenerated catalyst is divided into two portions: one portion flows through the regeneration pipe (19) back into the riser (22) while the other portion enters the cooler (16) through a pipe (15) and, after being cooled, flows through the regeneration pipe (17) back into the gasoline riser (2) for re-use. The

The pre-heated gasoline stock flows through a pipe (1) into the bottom section of the gasoline riser (2). The cooled regenerated catalyst flows through a regeneration pipe (17) into the bottom section of the riser (2). The reaction products flow into the second reaction section (34) of the new-type stock oil riser (22).

Meanwhile, the pre-rising medium flows through a pipe (20) into the bottom section of the new-type riser (22). The hot regenerated catalyst flows through the regeneration pipe (19) into the pre-rising section (a) of the riser (22), where it is subjected to the rising process by the pre-rising medium. The pre-heated conventional cracking stock flows through a pipe (21) into the riser (22), where it meets and reacts with the hot catalyst in the first reaction section (b) of the riser (22). The reaction products flow into the second reaction section (c) of the riser (22), where they have the secondary reaction with the reaction products coming from the gasoline riser (2).

The oil vapor and the regenerable catalyst flow through the outlet section (d) of the riser (22) and the horizontal pipe (e) into the precipitator (27). The oil vapor and the steam flow through a pipe (28) into the separation system (29). The gases and the gasoline products flow out of the device by a pipe (31). The diesel and heavier oil products exit the device by another pipe (30).

The regenerable catalyst enters the stripper (23) and, after being stripped by the steam coming from a pipe (24), flows through another pipe (25) into the regenerator (13). The regenerable catalyst is regenerated through the coke-burning process in the regenerator. The air comes into the regenerator through a pipe (14). The fume exits the regenerator through another pipe (12). The hot regenerated catalyst is divided into two portions: one portion flows through the regeneration pipe (19) back into the riser (22) while the other portion enters the cooler (16) through a pipe (15) and, after being cooled, flows through the regeneration pipe (17) back into the gasoline riser (2) for re-use. The disengaging air flow comes into the cooler through a pipe (18).

This Invention has several distinctive features:

1.   This Invention employs riser or fluid bed reactors in continuous reaction-

2. This Invention can process gasoline with 20-90wt% olefin and can use various types of catalysts.

3. This Invention can reduce the sulfur content in low-quality gasoline stock by 1/4-3/4 and reduce its nitrogen concentration to approximately 1ppm.

4. This Invention and the existing catalytic cracking unit can share the fractional system, the stripper, and the regenerator. The only additional equipment required is a gasoline stock riser or fluid bed reactor and a regenerated catalyst cooler.

Figures 1-5 illustrate, respectively, the process flow of the catalytic conversion as represented in Application Examples 1-5 for reducing the olefin content in gasoline. The numbers in the illustrations have the following designations: the numbers 1, 4, 8, 10, 11, 12, 14, 15, 18, 20, 21, 24, 28, 30, 31, 32, 33, and 34 refer to pipes; the number 2 refers to the gasoline riser reactor; the number 3 and 23 refer to strippers; the numbers 5 and 25 refer to regenerable catalyst pipes; the numbers 6 and 26 refer to distribution plates; the numbers 7 and 27 refer to precipitators (which may or may not be equipped with a fluid bed reactor); the numbers 9 and 29 refer to separation systems; the number 13 refers to the regenerator; the number 16 refers to the cooler; the numbers 17 and 19 refer to the regenerated catalyst pipes; and the number 22 refers to the stock oil riser reactor. Figure 6 illustrates the new-type riser reactor, wherein the letters a, b, c, d, and e refer to the pre-rising section, the first reaction section, the second reaction section, the outlet section, and the horizontal pipe respectively.

The following application examples offer more detailed descriptions of this Invention, but they are not the only available application of this Invention. Tables 1 and 2 list the properties of the catalysts and stock oils used in the application examples respectively. All the catalysts listed in Table 1 are produced by the Sinopec Qi Lu Catalyst Plant.

Application Example 1

This application example illustrates how to implement the catalytic conversion

Using Gasoline Stock A listed in Table 1 and the four different types of catalysts listed in Table 2, perform the olefin reduction test by means of a catalytic conversion process in continuous reaction-recycling operations in small-size fluid bed reactors. Mix the gasoline stock with high-temperature steam. Let the mixture enter a fluid bed reactor and have catalytic conversion reactions with the catalyst upon contact under the following reaction conditions: the temperature at 300°C, the pressure at the reactor head at 0.2megaPa, the feeding weight-space velocity at 4 hours^{-1}, the catalyst/gasoline ratio at 6:1, and the water/gasoline ratio at 0.03:1. The reaction products, the steam, and the regenerable catalyst are separated in a precipitator. The reaction products are separated into gas products and liquid products. The regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the regenerated catalyst is cooled and recycled to the reactor for re-use. Table 3 lists the test conditions, the results, and the gasoline properties.

As is shown in Table 3, different types of catalysts have different effects on the catalytic conversion of the gasoline stock. The processed gasoline contains 42-55wt% isomerized alkane, 25-27wt% arene, and only 8.5-18.5wt% olefin. The sulfur content in the gasoline has been reduced to 40-125ppm, and the nitrogen content has been reduced to 0.4-0.85ppm.

Application Example 2

This application example illustrates how to implement the catalytic conversion process as listed in this Invention to reduce the olefin content in gasoline when using gasoline stocks of different olefin content levels in small-size fluid bed reactors.

Using the four different types of gasoline stocks listed in Table 1 and Catalyst A listed in Table 2, perform the olefin reduction test by means of a catalytic conversion process in continuous reaction-recycling operations in small-size fluid bed reactors. Mix the gasoline stock with high-temperature steam. Let the mixture enter a fluid bed reactor

reaction products are separated into gas products and liquid products. The regenerable

catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the regenerated catalyst is cooled and recycled to the reactor for re-use.

Table 4 lists the test conditions, the results, and the gasoline properties. As is shown in Table 4, the resultant gasoline converted from gasoline stocks of different olefin content levels contains 54-65wt% isomerized alkane, 6.5-56.0wt% arene, and only 3.5-17.0wt% olefin. The sulfur content in the gasoline has been reduced to 40-578ppm, and the nitrogen content has been reduced to 0.4-1.6ppm. Gasoline stocks of higher olefin content levels yield gasoline of higher isomerized alkane content levels through the catalytic conversion process.

Application Example 3

This application example illustrates how to implement the catalytic conversion process as listed in this Invention to reduce the olefin content in gasoline under different operation conditions in small-size fluid bed reactors.

Using Gasoline Stock A listed in Table 1 and Catalyst A listed in Table 2, perform the olefin reduction test by means of a catalytic conversion process in continuous reaction-recycling operations in small-size fluid bed reactors. Mix the gasoline stock with high-temperature steam. Let the mixture enter a fluid bed reactor and have catalytic conversion reactions with the catalyst upon contact under the following reaction conditions: the temperature at 250-450°C, the pressure at the reactor head at 0.2megaPa the feeding weight-space velocity at 4-10 hours^{-1}, the catalyst/gasoline ratio at 3-8:1, and the water/gasoline ratio at 0.03-0.05:1. The reaction products, the steam, and the regenerable catalyst are separated in a precipitator. The reaction products are separated into gas products and liquid products. The regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the

As is shown in Table 5, different operation conditions have different effects on the catalytic conversion of the gasoline stock. The processed gasoline contains 52.5-57.0wt% isomerized alkane, 25.0-26.5wt% arene, and only 5.5-9.5wt% olefin. The sulfur content in the gasoline has been reduced to 36-46ppm, and the nitrogen content has been reduced to 0.3-0.41ppm.

Application Example 4

This application example illustrates how to implement the catalytic conversion process as listed in this Invention to reduce the olefin content in olefin-rich gasoline in medium-size riser reactors. The results of this test can be used to simulate the gasoline risers in double-riser reactors.

Using Gasoline Stock B listed in Table 1 and Catalyst C listed in Table 2, perforn the olefin, sulfur, and nitrogen reduction test by means of a catalytic conversion process in continuous reaction-recycling operations in medium-size riser reactors. Mix the gasoline stock with high-temperature steam. Let the mixture enter the bottom section of the riser and have catalytic conversion reactions with the catalyst upon contact under the following reaction conditions: the temperature at 550°C, the pressure at the reactor head at 0.2megaPa, the feeding weight-space velocity at 50 hours^{-1}, the catalyst/gasoline ratio at 6:1, and the water/gasoline ratio at 0.03:1. The reaction products, the steam, and the regenerable catalyst are separated in a precipitator. The reaction products are separated into gas products and liquid products. The regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the regenerated catalyst is cooled and recycled to the reactor for re-use. Table 6 lists the test conditions and results. Table 7 lists the gasoline properties.

As is shown in Table 7, the processed gasoline contains 40.32wt% isomerized alkane, 30.86wt% arene, and only 16.49wt% olefin. The sulfur content in the gasoline has been reduced to 97ppm, and the nitrogen content has been reduced to 0.76ppm. In

Application Example 5

This application example illustrates how to implement the catalytic conversion process as listed in this Invention to reduce the olefin, sulfur, and nitrogen contents when using lighter narrow cut fractions and heavier narrow cut fractions of olefin-rich gasoline and having them flow into the bottom section and the upper mid section of a medium-size riser reactor.

Using the lighter gasoline fraction C and the heavier gasoline fraction D listed in Table 1 and Catalyst A listed in Table 2, perform the olefin, sulfur, and nitrogen reduction test by means of a catalytic conversion process in continuous reaction recycling operations in medium-size riser reactors. Mix the lighter gasoline fraction C with high-temperature steam. Let the mixture enter the bottom section of the riser and have catalytic conversion reactions with the catalyst upon contact at 300°C. Meanwhile, let the heavier gasoline fraction D enter the mid section of the riser and have catalytic conversion reactions with the catalyst upon contact at 400°C. The reaction conditions are as follows: the pressure at the reactor head at 0.2megaPa, the feeding weight-space velocity at 50-100 hours^{-1}, the catalyst/gasoline ratio at 6:1, and the water/gasoline ratio at 0.03:1. The reaction products, the steam, and the regenerable catalyst are separated in a precipitator. The reaction products are separated into gas products and liquid products. The regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the regenerated catalyst is cooled and recycled to the reactor for re-use. Table 6 lists the test conditions and results. Table 7 lists the gasoline properties.

As is shown in Table 6, the gas yield is 6.99wt%, the diesel yield is 4.53wt%, and the gasoline yield is 86.78wt%. As is shown in Table 7, the processed gasoline contains 51.25wt% isomerized alkane, 26.96wt% arene, and only 8.59wt% olefin. The sulfur content in the gasoline has been reduced to 86ppm, and the nitrogen content has been reduced to 0.65ppm.

This application example illustrates how to implement the catalytic conversion process as listed in this Invention to reduce the olefin content in gasoline in small-size fixed fluid bed reactors.

Using Gasoline Stock C listed in Table 1 and Catalyst A listed in Table 2, perform the olefin reduction test by means of a catalytic conversion process in continuous reaction-recycling operations in small-size fluid bed reactors. Mix the gasoline stock with high-temperature steam. Let the mixture enter a fluid bed reactor and have catalytic conversion reactions with the catalyst upon contact under the following reaction conditions: the temperature at 150-300°C, the pressure at the reactor head at 0.2megaPa, the feeding weight-space velocity at 4 hours^{-1}, the catalyst/gasoline ratio at 6:1, and the water/gasoline ratio at 0-0.03:1. The reaction products, the steam, and the regenerable catalyst are separated in a precipitator. The reaction products are separated into gas products and liquid products. The regenerable catalyst goes into a stripper and is steam stripped of the hydrocarbons clinging to the catalyst. The stripped catalyst is then sent to a regenerator to be regenerated in the presence of heated air. Finally, the regenerated catalyst is cooled and recycled to the reactor for re-use. Table 8 lists the test conditions, the results, and the gasoline properties.

As is shown in Table 8, the processed gasoline contains 61-65wt% isomerized alkane, 6.5-7.0wt% arene, and only 15.5-20.0wt% olefin. The sulfur content in the gasoline has been reduced to 100-140ppm, and the nitrogen content has been reduced to 0.65-1.10ppm.

Table 1

Gasoline Stock No.	A	B	C	D
Density (20°C), kg/m^3	743.0	727.1	653.1	786.4
Octane Rating				
RON	91.3	92.1	93.2	88.6
MON	79.5	79.8	81.5	78.4
Sulfur, ppm	200	2035.5	376.7	2892.2
Nitrogen, ppm	30	151.3	14.3	93.0
Carbon, wt%	86.80	86.54	85.26	86.29
Hydrogen, wt%	13.03	13.26	14.52	12.99
Fractional Process, °C				
Preliminary Fraction	49	44	44	90
10%	63	59	48	91
30%	82	78	53	120
50%	106	104	60	153
70%	140	133	65	173
90%	176	166	75	186
Final Fraction	195	200	90	202
Group Composition, wt%				
Alkane	29.64	26.94	19.04	25.00
Normal Alkane	5.29	4.88	4.98	5.12
Isomerized Alkane	25.35	22.06	14.06	19.88
Naphthenic Hydrocarbon	8.25	7.16	5.17	12.30
Olefin	37.79	47.65	69.64	13.62
Arene	24.32	18.25	6.15	49.08

Table 2

Catalyst No.	A	B	C	D
Brand Name	CRC-1	RHZ-200	ZCM-7	RAG-1
Zeolite Type	REY	REHY	USY	REY-USY-ZRF
Chemical Composition, wt%				
Alumina	26.5	33.0	46.4	44.6
Sodium Oxide	0.19	0.29	0.22	0.13
Ferric Oxide	0.09	1.1	0.32	--
Apparent Density, kg/m^3	450	560	690	620
Pore Volume, ml/g	0.41	0.25	0.38	0.36
Specific Surface Area, m^2/g	132	92	164	232
Abrasion Index, wt%hours^{-1}	4.2	3.2	--	2.5
Sieve Composition, wt%				
0-40 microns	7.3	15.2	4.8	13.1
40-80 microns	43.7	55.1	47.9	54.9
> 80 microns	49.0	29.7	47.3	32.0

Table 3

Catalysts		A	B	C	D
Reaction Temperature, °C		300	300	300	300
Weight-Space Velocity, hours^{-1}		4	4	4	4
Catalyst/Gasoline Ratio		6	6	6	6
Water/Gasoline Ratio		0.03	0.03	0.03	0.03
Product Distribution, wt%					
Dry Gas		1.36	0.87	0.65	0.56
Liquefied Gas		3.87	4.69	4.76	4.93
Gasoline		85.97	86.85	88.70	89.95
Light Diesel		4.37	3.98	3.01	2.36
Heavy Diesel		2.43	2.02	1.63	1.23
Coke		1.98	1.56	1.23	0.93
Loss		0.02	0.03	0.02	0.04
Gasoline Properties	Stock Properties				
RON	91.3	88.2	89.3	90.2	90.5
MON	79.5	80.1	80.0	79.8	79.8
Sulfur, ppm	200	40	65	102	125
Nitrogen, ppm	30	0.4	0.7	0.76	0.85
Arene, wt%	24.32	25.20	25.00	25.30	26.56
Olefin, wt%	37.79	8.70	10.87	14.98	18.39
Alkane, wt%	29.64	59.01	56.92	52.29	47.19
Normal Alkane	5.29	5.01	5.05	4.98	4.86
Isomerized Alkane	25.35	54.0	51.87	47.31	42.33
Naphthenic Hydrocarbon, wt%	8.25	7.09	7.21	7.43	7.86

Table 4

Stock Oil	A	B	C	D
Reaction Temperature, °C	300	300	300	300
Weight-Space Velocity, hours[-1]	4	4	4	4
Catalyst/Gasoline Ratio	6	6	6	6
Water/Gasoline Ratio	0.03	0.03	0.03	0.03
Product Distribution, wt%				
Dry Gas	1.36	1.57	0.73	1.33
Liquefied Gas	3.87	4.64	6.05	7.14
Gasoline	85.97	84.11	86.39	81.83
Light Diesel	4.37	5.04	3.33	4.32
Heavy Diesel	2.43	2.61	1.36	2.04
Coke	1.98	2.02	2.12	3.14
Loss	0.02	0.01	0.02	0.20
Gasoline Properties				
RON	88.2	90.8	90.2	89.4
MON	80.1	81.0	81.2	78.6
Sulfur, ppm	40	402	108.5	578
Nitrogen, ppm	0.4	1.0	0.83	1.6
Arene, wt%	25.20	19.20	6.52	55.18
Olefin, wt%	8.70	10.92	16.34	3.89
Alkane, wt%	59.01	62.72	70.02	27.48
Normal Alkane	5.01	4.95	5.20	5.22
Isomerized Alkane	54.0	57.77	64.82	22.26
Naphthenic Hydrocarbon, wt%	7.09	7.16	7.12	13.45

Table 5

Operation Conditions							
Reaction Temperature, °C		300	300	300	300	250	450
Weight-Space Velocity, hours^{-1}		8	4	4	4	4	10
Catalyst/Gasoline Ratio		3	6	8	6	6	6
Water/Gasoline Ratio		0.03	0.03	0.03	0.05	0.03	0.05
Product Distribution, wt%							
Dry Gas		1.12	1.36	1.56	1.13	0.56	3.41
Liquefied Gas		3.55	3.87	4.32	3.53	1.87	5.43
Gasoline		87.11	85.97	84.54	86.61	87.02	82.59
Light Diesel		4.01	4.37	4.63	4.18	5.47	3.24
Heavy Diesel		2.11	2.43	2.75	2.33	2.86	1.85
Coke		1.86	1.98	2.02	1.95	1.96	2.93
Loss		0.24	0.02	0.18	0.27	0.26	0.29
Gasoline Properties	Stock Properties						
RON	91.3	89.0	88.2	87.8	89.0	87.6	88.8
MON	79.5	80.0	80.1	80.1	79.9	80.0	80.0
Sulfur, ppm	200	46	40	36	42	36	36
Nitrogen, ppm	30	0.41	0.4	0.3	0.4	0.3	0.3
Arene, wt%	24.32	25.12	25.20	26.45	25.01	25.03	26.02
Olefin, wt%	37.79	9.32	8.70	6.67	9.13	5.98	9.09
Alkane, wt%	29.64	58.45	59.01	59.80	58.76	61.84	57.30
Normal Alkane	5.29	5.07	5.01	5.11	5.07	5.03	4.98
Isomerized Alkane	25.35	53.38	54.0	54.69	53.69	56.81	52.32
Naphthenic Hydrocarbon, wt%	8.25	7.11	7.09	7.08	7.10	7.15	7.09

Table 6

	Application Example 4	Application Example 5
Operation Conditions		
Reaction Temperature, °C	550	300/400*
Weight-Space Velocity, hours[-1]	50	50/100*
Catalyst/Gasoline Ratio	6.0	6.0
Water/Gasoline Ratio	0.03	0.03
Product Distribution, wt%		
Dry Gas	0.66	1.01
Liquefied Gas	3.55	3.87
Including Propylene	3.83	
Isobutane	5.58	
Gasoline	75.62	86.78
Light Diesel	3.75	3.52
Heavy Diesel	2.53	1.01
Coke	2.43	1.65
Loss	0.41	0.05

* The value preceding the slash mark (/) refers to the reaction condition in the bottom section of the gasoline riser and the value following the slash mark (/) refers to the reaction condition in the mid section of the gasoline riser.

Table 7

	Stock A	Application Example 4	Stock C	Stock D	Application Example 5
Density (20°C), kg/m^3	743.0	741.8	653.1	786.4	746.7
Octane Rating					
RON	91.3	90.0	93.2	88.6	91.0
MON	79.5	80.8	81.5	78.4	81.2
Sulfur, ppm	200	97	376.7	2892.2	86
Nitrogen, ppm	30	0.76	14.3	93.0	0.65
Carbon, wt%	86.80	86.71	85.26	86.29	86.43
Hydrogen, wt%	13.03	13.22	14.52	12.99	13.42
Fractional Process, °C					
Preliminary Fraction	49	49	44	90	46
10%	63	75	48	91	72
30%	82	98	53	120	90
50%	106	121	60	153	118
70%	140	147	65	173	143
90%	176	178	75	186	175
Final Fraction	195	202	90	202	196
Group Composition, wt%					
Arene	24.32	30.86	6.15	49.08	26.98
Olefin	37.79	16.49	69.64	13.62	8.59
Alkane	29.64	45.34	19.04	25.00	56.57
Normal Alkane	5.29	5.02	4.98	5.12	5.32
Isomerized Alkane	25.35	40.32	14.06	19.88	51.25
Naphthenic Hydrocarbon	8.25	7.31	5.17	12.30	7.86

Table 8

Operation Conditions		150	200	250	300
Reaction Temperature, °C		150	200	250	300
Weight-Space Velocity, hours^{-1}		4	4	4	4
Catalyst/Gasoline Ratio		6	6	8	6
Water/Gasoline Ratio		0	0.02	0.02	0.03
Product Distribution, wt%					
Dry Gas		0.86	0.51	0.64	0.73
Liquefied Gas		2.04	2.68	4.30	6.05
Gasoline		93.24	93.23	90.02	86.39
Light Diesel		1.65	1.52	2.46	3.33
Heavy Diesel		0.76	0.73	1.09	1.36
Coke		1.33	1.30	1.40	2.12
Loss		0.12	0.03	0.09	0.02
Gasoline Properties	Stock Properties				
RON	93.2	88.2	89.8	90.0	90.2
MON	81.5	81.2	81.5	81.5	81.5
Sulfur, ppm	376.7	106.5	137.2	101.3	108.5
Nitrogen, ppm	14.3	0.65	1.10	0.65	0.83
Arene, wt%	6.15	6.55	6.66	6.50	6.52
Olefin, wt%	69.64	16.70	19.53	15.98	16.34
Alkane, wt%	19.04	68.74	66.49	70.30	70.02
Normal Alkane	4.98	5.30	5.33	5.17	5.20
Isomerized Alkane	14.06	63.44	61.16	65.13	64.82
Naphthenic Hydrocarbon, wt%	5.17	8.01	7.32	7.22	7.12